

Anderson-Mott Transition Driven by Spin Disorder: Spin Glass Transition and Magnetotransport in Amorphous GdSi

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A zero temperature Anderson-Mott transition driven by spin disorder can be ‘tuned’ by an applied magnetic field to achieve colossal magnetoconductance. Usually this is not possible since spin disorder by itself cannot localise a high density electron system. However, the presence of strong structural disorder can realise this situation, self consistently generating a disordered magnetic ground state. We explore such a model, constructed to understand amorphous GdSi, and highlight the emergence of a spin glass phase, Anderson-Mott signatures in transport and tunneling spectra, and unusual magneto-optical conductivity. We solve a disordered strong coupling fermion-spin-lattice problem essentially exactly on finite systems, and account for all the qualitative features observed in magnetism, transport, and the optical spectra in this system.

The ‘Anderson-Mott’ insulator-metal transition (IMT) in disordered interacting systems [1,2] and the physics of ‘colossal magnetoresistance’ (CMR) in, for example, the manganese oxides [3] are topics of great current interest. Even though the “ultimate” example of CMR would be a magnetic field driven, zero temperature, insulator-metal transition, such a scenario had not been realised experimentally till recently. Most observations of CMR are at finite temperature, across a ferromagnet to paramagnet transition [3–5]. *Canonical ‘Anderson-Mott’ systems do not show large magnetoconductance (MC) and the standard CMR systems do not involve localisation physics.* These two fields of research have evolved independently.

Experiments [6–10] on amorphous a -GdSi reveal that the presence of doped magnetic moments in a strongly disordered system can combine features of the standard doping driven IMT in amorphous systems [11] with the physics of field driven IMT and CMR. The magnetic ground state in such a system is a spin glass. There are distinct signatures of electron correlation in the conductivity and tunneling spectra, and huge transfer of optical spectral weight on application of a magnetic field. The experimental observations, discussed below, cannot be understood within the standard scenarios [1–5] developed for IMT and CMR and require an independent and comprehensive framework. Our main achievement in this paper is (i) to provide the first understanding of the unique properties of this system, and (ii) demonstrate a many body technique that allows controlled approximations in a strongly disordered interacting system.

The measurements have been made on a -GdSi and simultaneously on the non-magnetic analog a -YSi. (i) Both Y_xSi_{1-x} and Gd_xSi_{1-x} show an IMT [6] as the doping, x , is increased beyond a critical value, x_c . The critical doping $x_c \sim 14\%$ in YSi and $x_c \sim 15\%$ in GdSi. (ii) For $x \lesssim x_c$ in GdSi, a magnetic field, $h_c(x)$, can actually drive an insulating sample metallic. YSi samples

show weak *positive* magnetoresistance. (iii) The density of states (DOS) at the Fermi level, $N(0)$, in GdSi, probed through tunneling conductance measurement [7], grows as $(h-h_c)^2$ across the IMT, while σ_{dc} increases as $(h-h_c)$. (iv) The optical conductivity in GdSi shows large transfer of spectral weight [8] to low frequency from $\omega \gtrsim 0.1$ eV on application of a field of a few Tesla. Transfer of spectral weight also occurs on raising temperature, and this is seen in both GdSi and YSi. (v) The low field a.c susceptibility in GdSi reveals [9] that the magnetic degrees of freedom freeze into a spin glass state at low temperature. The freezing temperature, T_f , increases from ~ 1 K at $x = 0.04$ to $T_f \sim 6$ K at $x \sim 0.20$. The Y doped samples are *diamagnetic*. (vi) The ‘effective moment’ inferred [9] from $\chi(T)$ differs from the expected value for Gd, and the high temperature magnetic specific heat [10] per doped Gd is almost 50% larger than $\log(2S + 1)$.

Observation (i) above is standard in disordered systems, (ii) – (iv) would be expected in CMR materials, and (v) – (vi) seem to be unique to the combination.

A ‘first principles’ model for amorphous GdSi will have to consider an underlying ‘random’ structure in which a fraction x of the sites are occupied by Gd atoms and $(1-x)$ by Si atoms. The Gd and Si atoms have different orbital structure so a complicated set of inter-orbital, intersite hopping possibilities need to be considered. We try to retain the essential features in the following, simpler, one band model:

$$H = -t \sum_{\langle ij \rangle, \sigma} c_{i\sigma}^\dagger c_{j\sigma} + \sum_{i\sigma} (\epsilon_i - \mu) n_{i\sigma} - J' \sum_{\nu} \sigma_{\nu} \cdot S_{\nu} - \lambda \sum_{\nu} n_{\nu} x_{\nu} + \frac{1}{2} K \sum_{\nu} x_{\nu}^2 + H_{Coul} \quad (1)$$

We use a tight binding model with uniform hopping t , and an on site potential ϵ_i uniformly distributed between $\pm \Delta/2$. The sites labelled ‘ ν ’ are a fraction x of the lattice corresponding to the dopant (Y/Gd) locations. The

electron-spin coupling is J' , \mathbf{S}_i are the $S = 7/2$ Gd spins, and $\sigma_{i\mu} = \sum_{\alpha\beta} c_{i\alpha}^\dagger \tau_\mu^{\alpha\beta} c_{i\beta}$, where τ_μ are the Pauli matrices. The x_i are local displacement variables (bond distortions [12]) coupled to the electron density via λ , and the structural stiffness is K . H_{Coul} would include Hubbard and long range Coulomb interactions.

The width of the impurity level distribution (Δ) in YSi/GdSi has been estimated [8] to be ~ 200 meV, and the ‘polaron binding energy’ $g = \lambda^2/K \sim 30$ meV. The existence of lattice polaron effects in amorphous semiconductors had been argued early on by Anderson [12], and has been revived now [8] in the context of doped a -Si. We think the diamagnetism in YSi confirms bipolaronic effects, but these lattice effects are probably not very important in GdSi. There is no simple estimate of the ‘effective hopping amplitude’ to be used in a single band approximation. However, calculations on the Anderson model indicate [13] that we need $\Delta/t \approx 14$ to localise 10% of the electronic states in the band. This suggests a rather small effective hopping amplitude ~ 200 K, if Δ is 200 meV. The electron-spin coupling $J'S$ (called J' from now on), arising out of the $d-f$ coupling in Gd, is large. It is estimated to be ~ 0.9 eV from photoemission measurements [14] on Gd, but would be somewhat smaller in the effective one band description that we are using. Although the parameter values have some uncertainty it is clear that $\Delta, J' \gg t$. We use $\Delta/t = 11$, $J'/t = 4$, $g/t = 0.5$, roughly consistent with the experimental estimates. The electron-phonon and electron-spin coupling are operative only at the dopant sites. We distribute the ‘impurities’ (Y or Gd in the Si host) into Nx sites with the lowest potential in any given realisation $\{\epsilon_i\}$. This ensures that at low dopant concentration, the electrons are trapped near the dopant sites. We measure all energies in units of t , and finally assume $t \approx 500$ K for comparing our energy scales with the data.

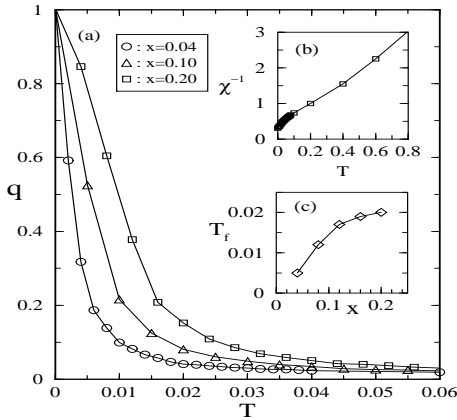


FIG. 1. (a). Order parameter for freezing, $q(T)$ (see text), within our effective magnetic model. Insets: (b). The inverse susceptibility $\chi^{-1}(T)$ over a large temperature range. (c). Freezing temperature $T_f(x)$. Simulation on 10^3 lattices.

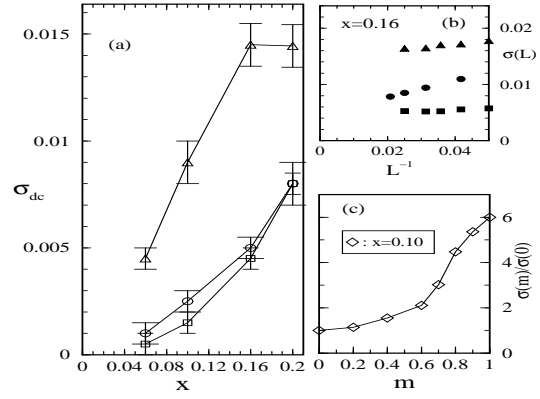


FIG. 2. (a): D.C. conductivity, at $T = 0$. Model for: YSi (circles), GdSi in the spin glass phase (squares), and GdSi in the fully polarised phase (triangles). (b): $L \rightarrow \infty$ extrapolation to construct σ_{dc} (see text). (c): GdSi: dependence of σ_{dc} , at $x = 0.10$ and $T = 0$, on magnetisation.

Our principal results using the Hamiltonian above are the following (i) the magnetic ground state in the low doping region is a spin glass with T_f having overall scale $\sim t^2/(J' + \Delta)$ and following the experimental doping dependence (Fig.1), (ii) there is a metal-insulator transition for both GdSi and YSi with decreasing x , with $x_{Gd}^c \gtrsim x_Y^c$, and a field driven IMT and CMR for GdSi (Fig.2), (iii) there is large transfer of spectral weight from high to low frequency (Fig.4) in $\sigma(\omega)$, driven by an applied magnetic field in GdSi. We explain our scheme of calculation next and then discuss these results in detail.

Since the Hamiltonian involves $\Delta/t, J'/t \gg 1$ and $g \sim \mathcal{O}(t)$ none of these couplings can be handled perturbatively. To study the properties of this model within a controlled approximation we use a finite size combination of Monte-Carlo and exact diagonalisation [15] (MC+ED). This approach exactly handles the strong disorder, but treats the spin and lattice variables as ‘classical’. Since we have $2S \gg 1$, the ‘classical’ spin limit should be a reasonable starting point. At strong disorder, the leading effect of phonons should also be accessible classically.

If we ignore H_{coul} to start with, H represents non-interacting fermions coupled to classical variables \mathbf{S}_i and x_i , in addition to the random potential ϵ_i . The ϵ_i are ‘quenched’ variables while the spin and lattice degrees of freedom are ‘annealed’, with the distribution $P\{x, S\} = Z^{-1} \text{Tr} e^{-\beta H}$ where $Z = \int \mathcal{D}S \mathcal{D}x \text{Tr} e^{-\beta H}$ is the full partition function for a specific realisation of $\{\epsilon_i\}$.

The ‘exact’ MC+ED allows only small system sizes, $\mathcal{O}(100)$ sites, so the key step is to construct an approximate ‘effective Hamiltonian’ for the lattice and spin variables. Once the magnetic and phonon problem are self consistently solved, the $T = 0$ electron problem can be solved in the classical ground state $\{\mathbf{S}_i, x_i\}_0$, which itself depends on $\{\epsilon_i\}$, finally averaging over disorder.

Formally the magnetic effective Hamiltonian is $H_{eff}\{S\} = -\frac{1}{\beta} \log \int \mathcal{D}x \text{Tr} e^{-\beta H}$. The magnetic prob-

lem involves $J'/t \gg 1$, a dilute system (the spins occupy only a fraction x of sites), and strong disorder in the electron system. There is no perturbative expansion possible in J' but the large J' and $x \ll 1$ allows a simplification. In this limit the doped carriers are essentially localised at the magnetic sites, with the electron density falling off exponentially away from the sites. This generates a pairwise *antiferromagnetic coupling*, for $R_{ij} > 1$, with $J_{ij} \sim (t^2/J')e^{-R_{ij}/\lambda(J')}$, with $\lambda \propto 1/J'$. For neighbour-sites there is a ferromagnetic coupling $\sim \mathcal{O}(t)$.

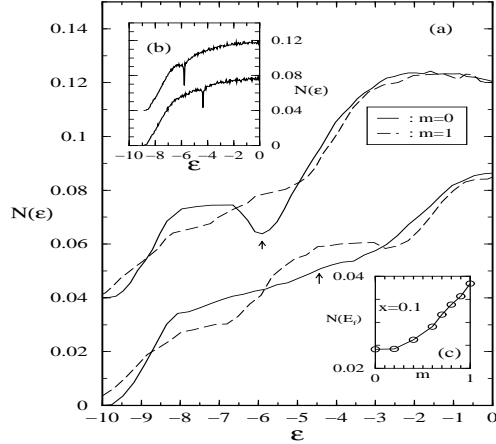


FIG. 3. (a). Density of states in the model for GdSi for two densities, $x = 0.1$ and $x = 0.2$, for random spins ($m = 0$) and polarised spins ($m = 1$). The curves are vertically shifted by 0.04. Fermi energy marked by arrows. (b). DOS in model for YSi, at $x = 0.1$ (above) and $x = 0.2$ (below). (c). DOS at ϵ_F in the model for GdSi, at $x = 0.1$, with increasing magnetisation.

In the disordered system, for a given separation R_{ij} , the bonds have a distribution. We construct the distributions $P(J, R_{ij})$ and study the model: $H_{eff}\{\mathbf{S}\} = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$. For a pair of moments located at \mathbf{R}_i and \mathbf{R}_j the J_{ij} is picked from $P(J, R_{ij})$. This ignores correlations between bonds in a specific $\{\epsilon_i\}$ realisation. We simulate the model for different dilutions, compute the order parameter for freezing, $q(T) = (xN)^{-1} \sum_i |\langle \mathbf{S}_i \rangle_T|$, and check that the structure factor has no peaks at any wavevector \mathbf{Q} . Fig.1 shows $q(T)$, alongwith $\chi(T)$ at $x = 0.2$, and the freezing temperature [16] $T_f(x)$. At $x = 0.2$ our $T_f \sim 10\text{K}$, while experimentally $T_f \sim 6\text{K}$. We have done the exact MC+ED simulation for 4^3 systems, with the same J', Δ and $x \sim 0.1$ and verified [17] that the system freezes into a spin glass, with T_f within 10% of our result here.

Having established the existence of a glassy state for the $\{\mathbf{S}_i\}$ we will simplify the remaining electron-phonon problem by assuming the spins to be frozen in an uncorrelated random manner.

The effective Hamiltonian for phonons is $H_{eff}\{x\} = -\frac{1}{\beta} \log \int \mathcal{D}S T r e^{-\beta H}$. At moderate g and strong disorder

there would be ‘frozen’ bond distortions in the ground state, and we cannot expand about the $x_i = 0$ state. To incorporate this effect we use the lowest order self-consistent expansion, *i.e.*, $H_{eff}\{x\} \approx \frac{1}{2} K \sum_i x_i^2 + \sum_i a_i x_i$ with $a_i = -\lambda \bar{n}_i$, where $\bar{n}_i = \langle n_i \rangle$, computed in the electronic ground state. The minimum of H_{eff} , *i.e.* the lattice distortion in the ground state, corresponds to $\bar{x}_i = (\lambda/K) \bar{n}_i$. The $T = 0$ problem now corresponds to electrons in the background of structural disorder, $\{\epsilon_i\}$, coupled to randomly oriented spins with coupling J' , and density coupled to a phonon field $x_i = (\lambda/K) \bar{n}_i$, *i.e.*,

$$H_{eff}^e = H_{kin} + \sum_i \epsilon_i n_i - J' \sum_\nu \sigma_\nu \cdot \mathbf{S}_\nu - g \sum_\nu \bar{n}_\nu n_\nu \quad (2)$$

We solve this problem through iterative ED. The transport and spectral properties of YSi correspond to $g = 0.5$ and $J' = 0$, while for GdSi $g = 0.5$ and $J' = 4$. The ED is done for a sequence of sizes $6 \times 6 \times L$, with $L = 24, 32, 40, 48$. Due to the finite size gaps the d.c. conductivity cannot be directly computed on finite systems. We use the Kubo-Greenwood formula to compute the integrated optical spectral weight $\sigma_{int}(\Delta\omega) = \int_0^{\Delta\omega} \sigma(\omega) d\omega$, disorder average, and invert to obtain the optical conductivity $\sigma(\omega)$. The extent of averaging varies from 400 – 100 realisations, decreasing with increasing L . We track $\sigma(\omega_{ref}; L)$ with $\omega_{ref} \propto L^{-1}$, setting $\omega_{ref} = 0.08$ at $L = 32$, and use $\sigma_{dc} = \lim_{L \rightarrow \infty} \sigma(\omega_{ref}, L)$. Fig.2(a) shows this ‘dc’ conductivity appropriate to YSi and GdSi. Our x_c are smaller than the experimental values and we have not fine tuned parameters to match the data. The σ are in units of $\pi e^2 / \hbar a_0$ and the typical values shown in Fig.2(a) are ~ 0.01 . For $a_0 \sim 2\text{\AA}$, $\sigma_{dc} \sim 400 (\Omega\text{cm})^{-1}$, roughly as in experiments [6]. Fig.2(b) shows the L dependence of $\sigma(\omega_{ref}; L)$, while Fig.2(c) shows the ‘magnetoconductance’. We have checked that the conductivity in a ‘spin glass’ background shows the same trend as for random spins and the numbers match within $\sim 20\%$.

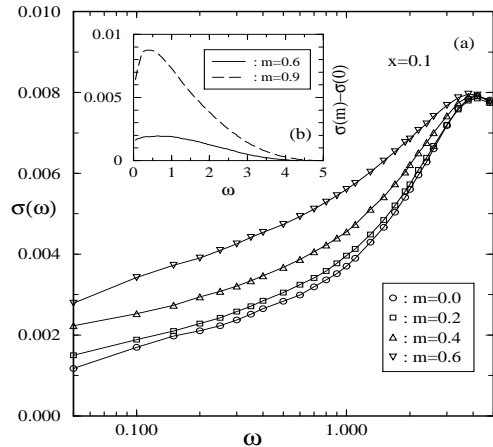


FIG. 4. Variation of optical conductivity in the model for GdSi, at $T = 0$, with degree of magnetisation. Note the log scale in frequency. Inset: magneto-optical conductivity.

To our knowledge there are no standard results on electron systems combining strong structural disorder *and* strong coupling to dilute magnetic moments. In the well studied opposite limit, $\Delta/t \gg 1$, $J'/t \ll 1$, spin flip scattering actually *weakens* [1] Anderson localisation, so large J'/t is crucial to enhanced localisation in GdSi. We have cross checked the trends in σ_{dc} by computing the averaged Greens function $G^{\sigma\sigma'}(\mathbf{r} - \mathbf{r}')$ at large Δ , with increasing J' . Using $TrG = \sum_{\sigma} G^{\sigma\sigma}(\mathbf{r} - \mathbf{r}')$ as the indicator of ‘delocalisation’, we find that at $|\mathbf{r} - \mathbf{r}'|/a_0 = 6$, TrG grows with J' upto $J' \sim 0.5$ and then falls rapidly [17]. It recovers quickly as the spins are polarised by an applied field, tracking the change in conductivity.

The DOS in GdSi, at low x , has a broad minimum at ϵ_F , Fig.3(a), since J' pulls down states to lower energy. This minimum is *not* related to the Altshuler-Aronov ‘correlation gap’ which would be a sharper feature [2] near ϵ_F (with effects of H_{coul} included). In YSi, the effect of phonons on the disordered background shows up as a sharp dip [1] in the DOS, Fig.3(b), since it generates a short range attractive interaction with $U_{eff} = -\lambda^2/K$.

We mimic the effect of finite magnetisation (m), in GdSi, by using a spin distribution with finite $\langle S_z \rangle$. Finite m leads to significant redistribution of weight in the DOS, due to the large J' , which should be visible in photoemission measurements. The conjunction of increased mobility, and increased DOS near ϵ_F , Fig.3(c), leads to the large changes observed in $\sigma(\omega)$, Fig.4. The ‘outer scale’ in $\sigma(\omega)$ is $\sim 5t \approx 0.25$ eV, as in the data [8].

There are certain experimental features for which the ‘Mott’ aspect is essential. These are principally the \sqrt{T} dependence in $\sigma_{dc}(T)$, the $\sqrt{\omega}$ correlation gap, the T driven spectral weight transfer in $\sigma(\omega)$, and the excess magnetic C_V . Most of these are generic correlation effects, well known in other amorphous systems [1,11], and unrelated to the magnetic character.

Let us re-emphasize the uniqueness of the system we study. Disorder, electron-spin coupling and electron-phonon interactions are features common, in some form, to a -GdSi, Anderson-Mott systems (NbSi, say) and the CMR manganites. The crucial differences are: (i) GdSi is a *strongly disordered ‘dilute’ magnetic system, with strong electron-spin coupling*. These features are essential to the spin glass behaviour and the consequent IMT and CMR. Electron-phonon interactions, even if present, are not crucial to the physics. (ii) Anderson-Mott systems are also *strongly disordered, but nominally non magnetic*. There are no remarkable magnetic field effects and the physics is controlled by disorder and electron correlations. (iii) Most CMR manganites are reasonable metals at low temperature, indicating *weak intrinsic disorder*. They have *strong electron-spin coupling* on a *periodic Mn lattice*, which, in contrast to ‘dilution’, promotes double exchange *ferromagnetism*. Electron-phonon (Jahn-Teller) interactions are important in these systems. The finite temperature IMT and CMR are related to multi-

phase coexistence [3] and not an Anderson transition. GdSi differs also from the diluted magnetic semiconductors in that the spin polaron concept [5,6] is not tenable in this high electron density system, due to strongly overlapping wavefunctions.

In conclusion, this is the first explanation of insulator-metal transition, CMR, spin glass freezing and optical properties of a -GdSi, bridging the gap between Anderson-Mott transition and CMR systems. Our results are based on an exact finite size calculation, handling strong disorder and interactions. The effect of Coulomb interactions is understood only qualitatively at the moment, and their inclusion would be the next step.

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